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Flow Processes in the Dry Regime: The Effect on Capillary Barrier Performance

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Abstract

Engineered capillary barriers typically consist of two layers of granular materials designed so that the contrast in sediment hydrologic properties and sloping interface retains infiltrating water in the upper layer. We report here on the results of two bench-top capillary barrier experiments, and associated modeling. We measured hydrologic parameters for both coarse materials using standard methods and found that the two materials had similar hydrologic properties despite being morphologically different (round, uniform vs. angular, non-uniform). The rounded sand provided a better functioning capillary barrier than the angular sand, but neither experiment could be characterized as a perfectly working capillary barrier. In both cases, more than 93% of the infiltrating water was successfully diverted from the lower layer, but infiltration into the underlying layer was observed in both systems. Based on this work, we believe that other non-continuum processes such as vapor diffusion and film flow contribute to the observed phenomena and are important aspects to consider with respect to capillary barrier design, as well as dry vadose zone processes in general. By applying a theoretical film flow equation representative of sediment surface geometries we were able to show that infiltration into the underlying sediment layer can be dominated by water film flow, a physical process that is typically not considered in numerical models of unsaturated flow in porous media.

Introduction

Computer simulation models have become essential tools for quantifying the physical processes associated with the near-surface environment in almost any scientific or engineering effort involving water resources. However, the reliable application of these computer models depends on the acquisition of representative physical soil properties and an accurate representation of the key underlying physical processes. It is ordinary practice to assume that the estimated van Genuchten (1980) or Brooks-Corey (1964) parameters adequately represent our materials in a hydrological sense. Nevertheless, material properties, such as grain morphology and surface roughness, can have a significant influence on the flow and transport properties of a porous media.

Under the study reported here, we present an example of the impact of grain morphology and surface roughness on predicting capillary barrier system efficiency, highlight inconsistencies in modeling, and discuss dry regime flow processes. Standard laboratory measurements of hydrologic properties were used to select two coarse gravels for the underlying layer in the two capillary barrier systems. Although van Genuchten parameters fitted to measured retention characteristics indicated that the two materials were hydrologically similar, they behaved differently under experimental conditions. We believe this is due to the significantly different surface areas and geometries of the two sediments.

By ignoring dry regime processes such as (1) film flow and (2) vapor diffusion, we were unable to accurately model water flow within the sediment. However, once film flow and vapor diffusion were included in the analyses, we could more successfully explain the observed behavior. The significance of the film flow component was found to be highly dependent on the surface roughness, with film infiltration velocities increasing with surface roughness and wettability. The concepts discussed are not limited to the conditions associated with a capillary barrier; they apply to many aspects of modeling unsaturated flow and transport in porous media in the dry regime.

Both film flow and vapor diffusion impact fluid flow in the dry regime, but they are typically neglected because their impact is negligible in most cases. However, under some circumstances these dry regime flow processes can dramatically impact the system behavior. Studies conducted by Hu et al. (2004) examined the impact of water content and thin water films on the movement of a solute through crushed tuff and found solute mobility to be a function of water film thickness and continuity. The presence of water films impacted the rate of diffusion within the system and the interconnectedness of pore space. In a similar capillary barrier system, and using the same materials as used here, Tidwell et al. (2003) observed equivalent breakthrough into coarse sediment, at an initial rate of 20% later slowing to 10% of the infiltrating fluid. The fluid breakthrough was attributed to water film flow and/or vapor transport.

Film flow and vapor diffusion contributions will manifest themselves in similar manner, following what is known as a Washburn relationship where $x \sim t^{1/2}$ (Bico et al. 2001), and are therefore difficult to decouple. The present work is focused on elucidating which mechanism is the dominant driver for these particular experiments. By using the theoretical relationships relating surface roughness to infiltration velocity developed by Hay et al. (2008) and qualitatively investigating theoretical vapor flux, we were able to isolate the primary mechanism for capillary barrier breach.

This work was initially motivated by the need to better understand the behavior of engineered capillary barriers made of backfill material. Capillary barrier design initially had the potential to be considered for high-level waste containment systems in which a capillary barrier would be established between the two different materials and water would be diverted away from the sensitive materials, such as vitrified glass or cement waste forms. Such barriers were considered in the preliminary design stage for the potential high-level nuclear waste site at Yucca Mountain, Nevada. The final design submitted with a license application in 2008 (http://www.nrc.gov/waste/hlw-disposal/yucca-lic-app.html) does not include this feature. However, capillary barrier systems are still being widely applied for instanceat landfills and as mine tailing covers, however with somewhat less rigorous design criteria.

The capillary barrier experiments presented here also provide an ideal example for analysis of discrepancies between ideal models and experimental system complexities; as the Richards-based model used here failed to predict the observed capillary barrier performance.

Film Flow

Film flow in unsaturated sediments has been described in many studies (Tokunaga 1997; Tokunaga et al. 2000). Tuller et al. (1999) and Tuller and Or (2001) pointed out that many models neglect film flow aspects by assuming that flow occurs only in full capillaries. Thus, the pressure potential is attributed to capillary forces only, while other potentially important adsorptive surface forces are ignored. This simplified model representation often provides satisfactory results for intermediate and highly saturated media, but, they tend to fail at low saturation (Nimmo et al. 1994).

To quantify the relative contribution of film flow in these systems we used knowledge of the sediment geometry, roughness and the theoretical expression of Hay et al. (2008). Wenzel (1936) first conceptualized the impact of surface roughness on wettablity and contact angle. Since then, extensive research has been conducted to quantify the effect of roughness on solid wetting and water film flow (Bico et al. 2001; Bico et al. 2002; Quere and Bico 2003; Martines et al. 2005; Hay et al. 2008). Hay et al. (2008) coupled surface geometries with the movement of water films to produce a conceptual model providing a pseudo sorptivity (or diffusion) term of water film infiltration velocities using only contact angles, surface tension, viscosity, and the sediment surface geometry.

Vapor Diffusion

DiCarlo et al. (1999) suggested vapor pressure gradients as a mechanism for enhanced movement of liquid and thus could account for differences in lateral spreading of moisture fingers observed in experiments. The vapor diffusion approach was based on the conservative assumption that the unsaturated hydraulic conductivity contribution to the sum of liquid and vapor transport (eq. 2 in DiCarlo et al. 1999) is extremely small at very low fluid saturations, and also that the vapor condenses to liquid behind the vapor front even in the absence of a temperature gradient. The latter assumption is supported by many studies showing that under equilibrium conditions vapor will condense, i.e., adsorb to porous materials, even though the porous material has the same temperature as the vapor. Parker (1986) explains that when a dry hydrophilic porous medium is placed in an atmosphere containing water vapor, isothermal water adsorption will increase with increasing vapor pressure until the pore space becomes fluid-filled. Similarly, Kutilek and Nielsen (1994) stated that the nature of soil water adsorption through hygroscopicity is completely different from the simple process of vapor condensation to its liquid phase. The adsorption phenomena are generally classified as being either physical (based on electrostatic and van der Waals attraction forces between the solid surface and water molecules) or chemical (based on rearrangement of electrons and consequent formation of strong chemical bonds) (Parker 1986; Nitao and Bear 1996). The adsorbed water layers coating the solid grains grow into films, and eventually adsorbed films in adjacent pore spaces will coalesce and form a continuous liquid phase in the pore. This process is generally referred to as capillary condensation (Derjaguin and Churaf 1974; Tuller et al. 1999). According to Easton and Machin (2000) there is no well-defined limit to the amount of vapor that can be absorbed for a wetting fluid; however, Tokunaga and Wan (2001) suggest that water films range in thickness from tens of nanometers to ~1μm. The adsorption of water vapor is aided by the vapor pressure deficiency that exists over a concave surface (air-water meniscus in a pore) compared to the vapor pressure over a free, flat water surface (Bear 1988).

In sharp contrast, Hu et al. (2004) report that Conca (1990) found that four sizes of tuff gravel remained dry after equilibrating for 70 days in a nearly 100% relative humidity. At the conclusion of the experiment only a 2.7% increase in intergranular water content was observed, indicating that the relative importance of vapor diffusion for water infiltration may be minimal. Vapor diffusion enhancement could contribute to the observed infiltration into the underlying layer in these experiments, but it is generally considered to only occur where a temperature gradient is present (Cass et al. 1984; Wildenschild and Roberts 2001). However, Webb and Ho (1998) reported both experimental and numerical modeling evidence of the enhancement of vapor diffusion in the absence of a thermal gradient. According to their study, the vapor density gradient, which drives the enhancement, can be established without the influence of a thermal gradient, for instance due to a vapor concentration gradient. In either case, enhancement across liquid bridges in the coarse material is a possible, but a fairly unlikely explanation, because at the saturation at which liquid islands develop in the coarse sand, the saturation level will also be sufficient to support capillary action, which is a much more efficient transport mechanism.

Thus the question remains, what is the relative impact of vapor diffusion or water film infiltration in unsaturated systems? It is likely that both processes are occurring simultaneously. The following experiments are targeted at understanding the importance of dry regime flow processes in a working capillary barrier system.

Experimental Design

The following sections describe the materials, surface area analysis, associated hydrologic parameters, and laboratory setup for the capillary barrier experiments.

Materials

Under our study, we conducted two experiments using commercially available Overton fine silica sand (#50–70 sieve) as the fine material and either 8/20 angular sand (#8–20 sieve) or 2/16 rounded sand (#2–16 sieve) as the coarse layer. The two experiments are referred to as Experiment 1 and 2, respectively. The coarse material for Experiment 1 was selected according to its utility as possible future backfill for the potential Yucca Mountain waste repository, thus a coarser crushed volcanic tuff was obtained from a supplier local to Yucca Mountain. For Experiment 2, sand with a similar grain size distribution was used, but we chose a material with a lower surface area and more rounded grains. The particular type of fine-grained material was selected because it would not filter into the coarse-grained material under dry conditions. See Table 1 for additional details about material properties.

Surface Area Analysis

Specific surface areas for the two coarse sands were measured using the Brunauer-Emmett-Teller (BET) (Brunauer et al. 1938) technique. The materials were out-gassed at room temperature to prevent sintering at high temperatures. Both total surface area and micropore area were measured in duplicate measurements and the results are listed (as averages of the measurements) in Table 1. As seen in Table 1, the 8/20 angular sand has 10 times higher total area than the 2/16 rounded sand. The 8/20 micropore area alone amounts to the total surface area for the 2/16 rounded sand.

Figure 1 shows microscopic photos of the two materials, and the difference between the two is quite noticeable. The 2/16 rounded sand consists of a very uniform quartz sand of very rounded grains, whereas the 8/20 angular sand is a volcanic tuff consisting of very angular grains containing various minerals and exhibiting intra-granular porosity.

Another difference between the two coarse materials is their grain morphology or angularity, and resulting pore shapes. Tuller et al. (1999) show in their Figure 1 that pore space geometry (the pore shape and angularity of grains) has a marked influence on the imbibition and drainage processes. During imbibition, the liquid-vapor interface in corners of angular pores grows with increasing potential (or capillary pressure) to the point of snap-off, whereas the round pores go from being completely empty to being full without the intermediate steps that occur in angular pores. Though both sediments in this system produced angular pores space, the angular 8/20 angular sand, produced more jagged edges and irregular pore space shape. Under drainage conditions, liquid displacement in cylindrical pores is piston-like, whereas in angular pores the liquid is displaced from the central region first leaving liquid in the corners. Subsequent increases in capillary pressure result in decreasing amounts of liquid in the corners (Tuller et al. 1999).

Hydrologic Parameters

The hydrologic characteristics of the materials were measured separately in smaller sample holders. The retention characteristics were measured using a quasi-static approach (Wildenschild et al. 1997) in a smaller pressure cell (7.6 cm diameter, 3.5 cm long). Saturated hydraulic conductivity was measured in a column (2.5 cm diameter, 28 cm long) using the constant head technique (eg. Klute 1986) for at least three different hydraulic gradients for each sample, and the unsaturated hydraulic conductivity was derived from the retention data. The resulting retention and unsaturated hydraulic conductivity curves for all materials used are illustrated in Figures 2 and 3. A non-linear least-squares optimization routine (RETC version 6.0 from the U.S. Salinity Laboratory) was used to fit van Genuchten (1980) parameters (listed in Table 2) to the curves. The two coarse materials have very similar hydrologic properties, except for a noticeable difference in residual saturation (Table 2), and a small difference in air entry pressure; but compared to the fine layer (Overton sand) they are very similar.

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Laboratory Setup – Capillary Barrier System

The capillary barrier experiments were carried out in a bench-top aluminum box (60.5 cm x 56.0 cm x 10 cm) with a Pyrex window on one side allowing easy detection of flow patterns (see Figure 4). Two 0.5-bar tensiometers were installed from the backside of the box (marked as squares in Figure 4). The tensiometers consisted of porous ceramic cups glued onto the backside of the box, and by testing were confirmed to have an air-entry value of at least 250 cm prior to use. Drainage out of the box was achieved by the use of two stainless steel, sintered rods installed from the backside immediately above the fine/coarse interface (circles in Figure 4). These drains had relatively low air-entry values (< 70 cm), but were highly permeable, such that water flow out of the box was not inhibited. They were connected to water-filled tubing, providing hanging column-based water phase tension to facilitate drainage under less than fully saturated conditions. Temperature variations during the experiments were measured with four thermocouples (1.02 mm diameter) placed in the box as shown in Figure 4 (stars).

The sandy materials were packed loosely in the box to simulate the emplacement of sediment using a conveyor belt such as would potentially be used at a large-scale waste site. As expected, the porosities of the materials varied slightly between the loose packing of the experimental box and the packing in the smaller pressure cells that were used for the hydrologic property measurements, the latter generally having lower porosities. We assume that the porosities for the experimental box were associated with the largest error due to inaccuracies of the material weights (loss during packing, etc.) and difficulty in precisely estimating the geometric boundaries of the individual layers. Before the materials were poured into the box, it was tilted to a 24-degree angle so that the fine/coarse interface was horizontal during packing. Table 3 lists relevant information from the experiment. An infiltration device was placed on top of the box to provide uniformly distributed infiltration over the entire surface area of the top of the box. A water-filled reservoir with 64 drips (0.25-mm inner diameter tubing and finger-tight fittings) was connected to a diaphragm pump. The pump rates were tested both before and after each experiment and are listed in Table 3. The average rate of infiltration into the underlying coarse layer for Experiment 1 was (29.8 – 27.8 ml/h) = 2.0 ml/h or 9.2 10^{-9} m/s, and for Experiment 2 only 0.1 ml/h (which is less than the standard error on the outflow rate) or 4.6 10^{-10} m/s. These numbers show that (assuming minimal or at least similar evaporation and other losses), the average infiltration into the coarse layer in the two experiments varied by a factor of 20. This calculation is a rough estimate, but provides a ballpark figure for the amount of fluid infiltration.

Results

The results of the capillary barrier experiments, associated numerical modeling and dry regime interpretation are described in the following sections.

Capillary Barrier Experiments

Infiltration and outflow rates for the two experiments are shown in Figure 5. Each box had an initial increase in outflow rate until steady-state flow conditions were established. In addition to infiltration and outflow rates, the average and standard deviations of the outflow rates are listed in Table 3. As seen in Figure 5, some water was retained in the box in Experiment 1 (Overton sand over 8/20 angular sand); the infiltration rate was higher than and exceeded one standard deviation of the average outflow rate. The drains diverted an average of 93.3% of the infiltrating water, which means that on average, the infiltration front moved at a rate of 6.7% of the applied inflow rate. This compares reasonably to the approximately 10% to 20% of the applied rate that Tidwell et al. (2003) found. In Experiment 2, using the 2/16 rounded sand as the coarse material, the infiltration rate was within a standard deviation of the outflow rate and the amount of water being withheld in the box was within measurement error. The amount of water diverted in this case was 99.7%.

The drain suction and tensiometer readings for the two experiments were monitored throughout the experiment. The air-entry value of the lower tensiometer (placed in the initially dry coarse material) was exceeded during the initial wetting procedure for both experiments and thus no readings are available for the capillary pressure in the coarse material during the experiments. After an initial adjustment period, the drain pressure was almost constant throughout the first experiment (43.2 cm +/-1.5 cm), whereas the capillary pressure measured at the upper tensiometer (in the fine material) varied slightly more (32.9 cm +/- 2.8 cm). The more notable fluctuations in capillary pressure in the box were closely correlated to temperature variations in the box, and are attributed to the temperature sensitivity of the transducers. In the second experiment the upper tensiometer was also fairly constant, apart from temperature-induced variations (35.7 cm +/- 2.8 cm). In this experiment the average drain suction was 46.3 cm +/- 2.1 cm, which was very similar to Experiment 1. A ceramic plate (initially intended for drainage) functioned as an additional tensiometer during part of this experiment and, after the initial non-steady-state wetting period, the measured capillary pressures were almost identical to the capillary pressures measured at the upper tensiometer. The upper tensiometer and the ceramic plate were located at a vertical distance of 21 cm from the base of the box and the fact that practically identical values were measured at both vertical locations indicates that flow had reached steady-state and was driven by gravity alone. The unsaturated hydraulic conductivity was thus equal to the flow rate through the box, providing us with measurement points on the unsaturated hydraulic conductivity curve for the Overton sand; these points are illustrated as individual points in Figure 3.

To better illustrate the flow patterns in the experiment, a dye tracer (Phenol Red) was sprinkled on the sand surface where it dissolved in the infiltrating water. In the first experiment it was added at the beginning of the experiment (initially dry sand), while in the second experiment it was added after 5 days when the wetting front had already reached the material interface. Phenol Red is a very conservative tracer with low adsorption capabilities. To document the tracer transport, images were periodically collected over the duration of the experiment. A time-lapse series of photographs for each experiment is shown in Figure 6 for Experiment 1 and 2, respectively. It is evident that in both experiments the water moved into the coarse layers and infiltrated faster (and further) into the 8/20 angular sand than into the 2/16 rounded sand. This is in agreement with the amounts of water diverted by the drains for the two experiments. Only 0.3 % of the infiltrated water was retained in the box for the second experiment (2/16 rounded sand as the coarse layer), whereas 6.7% was retained in the box for the first experiment (8/20 as the coarse layer). Tidwell et al. (2003) used materials that were identical to our Experiment 1 and derived very similar results: the barrier effectively diverted the majority of the infiltrating the water, and similar to our results, they found a slow and continuous infiltration into the coarse layer (see their Figure 2), the onset of which was noticed almost immediately after the wetting front reached the capillary interface. At the end of each experiment, an industrial vacuum was used to empty the box of sand. Successive layers were carefully removed and samples (approximately 25 ml) were collected. The samples were weighed and placed in a 105 ^{o}C oven over night and subsequently weighed again to determine the water saturation. The wetting front had progressed approximately 14 cm into the 8/20 angular sand, whereas only a narrow band of approximately 2 cm was significantly wetted in the 2/16 rounded sand. By analyzing the photographs of the experimental box taken at regular intervals and scaling the infiltrated distance to a feature of known length in the image, we estimated the infiltrated distances as a function of elapsed time as illustrated in Figure 7.

The infiltrated distance is plotted as a function of $t^{1/2}$, and it is obvious that the infiltration follows this relationship almost perfectly.

Numerical Modeling

We used the US1P module of NUFT (Non-isothermal Unsaturated-saturated Flow and Transport) (van Genuchten 1980; Nitao 1998) for the design simulations. This module solves the equations for single-phase unsaturated flow in porous media using the Richard's equation, which takes the form of:

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial x}\left[K(\psi)\frac{\partial \psi}{\partial x}\right]$$

in one dimension, where θ is the water content, K is the hydraulic conductivity, ψ is the matric potential and t is time. Matric potential-water content relationships were described using van Genuchten parameters where:

$$\theta(\psi) = \theta_r + \left(\frac{\theta_s - \theta_r}{1 + \left(\alpha|\psi|^n\right)^m}\right)$$

where θ_r is the residual water contentθ_s is the saturated water content, α is the inverse of the air entry pressure, *n* is a measure of the pore size distribution and *m* is equal to 1-(1/n). Also, NUFT uses saturations rather than moisture contents. Saturation, S, is defined as the moisture content, θ, divided by the porosity, ϕ. We assumed that $\theta_s = \phi$ (the porosity) in our simulations.

Before simulating the experiments described here, we compared the results of a capillary barrier simulation with results reported in Webb (1997) to gain confidence in our ability to model capillary barriers using NUFT. Webb's (1997) solution was selected due to its incorporation of characteristic curves and prior accurate prediction of capillary barrier performance. The NUFT simulations were in excellent agreement with Webb's results.

The two-dimensional model domain for the experiments is shown in Figure 8. The hydrologic properties used in the simulations are given in Table 4. In our numerical model, we describe the relationship between moisture content, capillary pressure (i.e., suction, head), and permeability using the van Genuchten (1980) and Mualem (1976) expressions (Mualem 1976; van Genuchten 1980).

Initially, the sands were assumed to be completely dry. The top boundary and the drain were held at constant head and saturation. The infiltration rate and drain suction used in the simulations are also given in Table 4. As a test, simulations for Experiment 1 were done with approximately double the grid resolution with no significant difference in model results. Simulations for Experiment 1 and 2 were run out to 33 days and 18 days, respectively, to match the actual length of the laboratory experiments.

Model Results

The saturation fields for Experiments 1 and 2 using the domain and grid shown in Figure 8 and the parameter values given in Table 4 are shown in Figure 9. As these figures clearly show, NUFT predicts successful performance of the barrier, and the complete absence of any wetting front migrating into the lower coarse material in both cases. However, a clear wetting front was observed for both cases in the actual experiments. The wetting front moved much faster and further into the 8/20 angular sand in the experiments, but because the two coarse materials have almost identical hydrological properties the model simulations predict nearly identical behavior for both experiments.

In attempting to capture the observed behavior with the numerical model, we adjusted various parameters to account for measurement error on the hydrologic properties. We used two principles to guide us with respect to which parameters we adjusted and by how much. The first was the likely bound of general parameter uncertainty. The second was the likely differences between the parameters determined from the drying curves we used to determine the values reported in Table 4 and the parameter values that we would have determined had we measured wetting curves rather than drying curves. In general, a wetting curve has a similar van Genuchten n-value but a greater α-value. For these experiments, wetting curves more likely represent the conditions in the coarse sand. For the fine sand, it is more difficult to know which curve better represents conditions in the system; most likely the most representative values are somewhere in between those for the two curves.

Infiltration into the underlying coarse material was observed as illustrated in Figure 10 when the α, m, porosity, and saturated conductivity values were altered to the values listed in Table 5. Manipulation was required to match the experimental results; K_s was halved and α doubled for the fine sand; K_s was halved and m and the porosity were altered to varying degrees for the coarse sands. Even though we were able to reproduce the results seen in the experiments by adjusting the hydrologic parameters, it seemed unwarranted to blindly venture into an exercise of pseudo parameter estimation when the likely cause was the lack of consideration of dry regime flow processes. This reasoning is also supported by the fact that the difference observed between the two experiments using the different coarse materials is not captured in a quantitative manner (see Figure 10) by this parameter estimation exercise.

Figure 11 illustrates the pressure-saturation curve for the measured hydraulic parameters (solid line) and the curves for the parameters required to obtain a model fit (dashed line). Clearly, the changes required in the α parameter of the fine sand dramatically impact the expected hydraulic properties of the soil, bringing the average pore throat radius significantly closer to that of the 8/20 angular and 2/16 rounded sand.

Dry Regime Experiments

Our modeling approach using NUFT yielded results that explained neither the breach of the barrier nor the differences between the two sediments; therefore, a number of alternative physical flow mechanisms must be considered. The numerical model predicts a perfectly working capillary barrier for both material combinations, unless the hydrologic property values are adjusted. This indicates that the wetting phenomena observed in the experiments are due to mechanisms not considered in these numerical simulations. Two different questions need to be addressed: 1) how do we account for the slow, but constant, wetting of the coarse layer, which happens near the fine/coarse layer interface in both experiments; and 2) why is there such a large variation in wetting front saturation level and depth for the two different coarse materials (despite the fact that they have very similar hydrologic properties)?

We believe that the primary cause for infiltration in the two systems was flow processes specific to the dry regime. The increased infiltration in Experiment 1 was primarily caused by significantly higher surface area and a higher natural wicking action from the sediments. To further explore the impact of dry regime processes, supplemental experiments and data were collected to examine the relative roles of film infiltration and vapor diffusion. Because the dry regime processes were most clearly demonstrated in Experiment 1, the 8/20 angular sand was used for additional measurements of vapor diffusion.

Vapor Diffusion Experiments

Vapor diffusion into sediment samples was monitored gravimetrically. Ten grams of coarse sediment was dried at 105º C and placed in a sealed closed container with an open reservoir of water. Relative humidity was monitored using a portable relative humidity sensor and maintained near 95%. The change in the mass of the sediment sample was monitored over a period of 10 days using an analytical balance accurate to +/-0.001 g.

Sediment Surface Characterization

Surface profiles of the 8/20 angular sand were obtained using a Nanonics Multiview 1000 atomic force microscope system, with a 70-micron scanner in non-contact mode with a non-contact probe. Image analysis using WxSM image processing software was used to obtain the average amplitude (δ) and wavelength (λ) of sediment surface features.

Results of Dry Regime Experiments

The dry regime experiments illustrated that vapor diffusion onto sediment surface in the absence of a gradient was minimal. Over a period of 10 days the mass change in sediment weight was within the experimental margin of error of the analytical balance. As mentioned previously, similar findings were reported by Conca (1990), who determined the vapor diffusion coefficient for tuff gravel samples to be 10^{-15} m^2/s (or 8.6 x 10^{-7} cm^2/day). At the saturated fine/coarse material interface in the experimental system discussed here, the relative humidity would be expected to be near 100%. Because the dry coarse material is overlain by wet fine sand, the relative humidity within the pore space of the coarse material would equilibrate within hours; therefore, we postulate that due to the lack of gradient in vapor concentration and low vapor diffusion coefficient, the impact of vapor diffusion on fluid movement is likely significantly less important than film flow and sediment wicking in this experimental system.

To close the book on the impact of vapor diffusion on the sustained infiltration into the coarse underlying layer, we also calculated vapor diffusion estimates based on DiCarlo's estimate (eq. 9 in DiCarlo et al. 1999) and based on a traditional Fickian approach applied to soils (Cass et al. 1984):

$$J_v = -a\alpha D\nabla\rho \qquad (0)$$

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where J_v is the mass flux density of water vapor (kg m^{-2} s^{-1}), D is the diffusion coefficient of water vapor in air (m^2 s^{-1}), $\nabla\rho$ is the water vapor density gradient (kg m^{-4}), a is the volumetric air-filled porosity (m^3 m^{-3}), and α is a dimensionless tortuosity factor generally assumed to be 0.66 for isothermal flow. The DiCarlo estimate is based on the conservative assumption that the unsaturated hydraulic conductivity contribution to the sum of liquid and vapor transport (eq. 2 in DiCarlo et al. 1999) is extremely small at very low fluid saturations, and also that the vapor condenses to liquid behind the vapor front even in the absence of a temperature gradient. The latter assumption is supported by many studies showing that vapor will condense, i.e., adsorb to porous materials, even though the porous material has the same temperature as the vapor. In Figure 12, the DiCarlo and Fickian vapor diffusion estimates are compared to the measured infiltration rates for Experiment 1. The cumulative vertical flux in these figures was calculated based on the saturation measured at the front edge of the wetting front over time which was 10.8%, and takes into account the ambient relative humidity of 85%. The driving gradient for vapor transport is the relative difference between the saturated and ambient humitdiites. For a relative humidity of 85%, the actual measured vertical flux for the 8/20 angular sand experiment is approximately 10 times higher than the Fickian estimate and 3 times higher than the DiCarlo estimate. If the relative humidity was closer to 100% as one would expect for the closed box we used in these experiments, the DiCarlo and Fickian estimates would be even lower due to the lack of driving gradient, and it is therefore not possible to explain the observed infiltration into the 8/20 angular sand based on vapor diffusion-based phenomena.

To confirm our hypotheses and theoretically verify the proposed dominance of film flow in the capillary barrier experiment, we applied the equation for film flow infiltration based on Wenzel wetting developed by Hay et al. (2008):

$$x_{h-p} = \left[\frac{\gamma\delta^2}{4\mu}\left(\frac{(2\delta+\lambda)cos\theta - \lambda sin\theta}{\lambda\delta}\right)\right]^{1/2} t^{1/2} = St^{1/2} \qquad (0)$$

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Including a negative capillary pressure at the coarse/fine interface, the above equation becomes:

$$x_{h-p} = \left[\frac{\delta^2}{4\mu}\left(-P+\gamma\frac{(2\delta+\lambda)cos\theta - \lambda sin\theta}{\lambda\delta}\right)\right]^{1/2} t^{1/2} = St^{1/2} \qquad (0)$$

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where x_{h-p} is infiltration distance (cm), γ is the surface tension (7.2x 10^{-2} N/m for water at 20 °C), δ is the average amplitude of surface features (nm), λ isthe average wavelength of surface features (nm), μ is the kinematic viscosity (1.02 x 10^{-3} Pa-s at 20 °C), t is time in seconds, P is the capillary pressure at the coarse/fine interface (-3432 Pa), and S is the sorptivity (or diffusion) term in m/s$^{1/2}$. Using this relationship and the measured quantities obtained from atomic force microscopy measurements given in Table 6, we obtained a theoretical sorptivity term of 7.7 cm/day$^{1/2}$ for the 8/20 angular sand and 5.7 cm/day$^{1/2}$ for the 2/16 rounded sand using a contact angle of 7 degrees. Adjusting for tortuosity, the vertical length of travel is 1/3 the actual travel distance (Dullien 1992), the estimated diffusion terms become 2.3 cm/day$^{1/2}$ and 1.7 cm/day, which matches the 8/20 angular (tuff) and 2/16 rounded experimental values of 2.54 cm/day$^{1/2}$ and 1.42 cm/day$^{1/2}$ quite well, see Table 7 for summary.

It should be noted that the sorptivity term varies as a function of the contact angle of the sediment, Figure 13 below illustrates the relationship. Between a range of zero and fifteen the calculated sorptivity term fluctuates by approximately 0.2 cm/day$^{1/2}$.

Discussion

The phenomena observed during our study are likely due to the combined effect of the two processes, as water film infiltration and vapor diffusion rarely occur separate from each other. In a similar experiment, Tidwell et al. (2003) proposed that because blue dye was absent in the infiltrated water (assuming that the large organic molecule would have been filtered out by films and evaporation) the observed slow, but constant, infiltration into the dry layer must be caused by the combined action of vapor diffusion and film flow. The initial rate of infiltration measured in the Tidwell et al. (2003) was 20 percent, slowing to 10 percent after 112 days. These rates are higher than the infiltration observed in these experiments, however that may be due to differences in experimental design.

The interaction of the two processes is schematically illustrated in Figure 14.

Initially, water vapor flows through the sand and some of the vapor adsorbs on the grains (Figure 14a-b), while the rest flows or diffuses through the open space between grains (Tzevelekos et al. 2000). At increasing vapor pressures water continues to be adsorbed onto the grains in multiple layers as long as sufficient water vapor is provided by vapor diffusion for this process to take place (Figure 14b-c). Over time, sufficiently thick films may form to facilitate flow (Figure 14c-d). Eventually the films on adjacent grains can coalesce via capillary condensation, initially in the finer pores, resulting in enhanced permeability and actual capillary action (Figure 14e-f). However, the timescales over which we are observing water moving through the coarse material indicate that vapor diffusion is not the dominant mechanism. As indicated by Conca (1990), and our own supporting experiment, vapor adsorption is minimal even under conditions of near 100% relative humidity. Vapor diffusion and capillary condensation onto similar crushed tuff material only accounted for a 2.4% change in intergranular moisture content. This is further supported by Jabro (2009), who found the rate of water vapor gain in larger soil aggregates to be minimal at room temperature.

In contrast, studies conducted by Bico and Quere and others (Bico et al. 2001; Bico et al. 2002; Quere 2002; Quere and Bico 2003; Ishino et al. 2004; Quere 2008) indicate that surface wetting resulting when water films flow over rough materials can occur nearly instantaneously. Martines et al. (2005) and Bico et al. (2001) illustrated that the movement of fluid across a surface of fabricated nanopatterns could be predicted by hemi-wicking theory, where fluid infiltration is a function of the wettability and surface geometry of a material. Using the Hay et al. (2008) expression relating sediment surface geometry to infiltration, we were able to estimate the sorptivity coefficient, which provided a reasonable approximation of infiltration into the underlying layer of the capillary barrier system in Experiment 1.

Differences in diversion capacity and barrier stability between Experiments 1 and 2 were likely due to the differences in surface properties of the 8/20 and 2/16 sands. As illustrated in Figure 1, and by the BET data, the surface of the 8/20 angular sand is significantly rougher than that of the

2/16 rounded sand. Experimental design and numerical modeling that do not consider the processes of vapor diffusion or film flow failed to predict the behavior of both capillary barrier systems. This is likely due to the exclusion of surface properties and the lack of consideration of dry regime processes in the numerical model.

Conclusion

We conducted two capillary barrier experiments using almost identical initial and boundary conditions, but using different underlying (coarse) materials. The coarse materials had very similar hydrologic properties, but were morphologically different. The rounded sand (2/16) provided a better functioning capillary barrier than the angular sand (8/20), but neither of the materials (in combination with the fine Overton sand) provided a perfectly working capillary barrier. Our experimental results and data analyses indicated that prediction of capillary barrier performance based on standard hydrologic property parameter measurements andRichard's equation is not always adequate for predicting detailed system behavior.

Our numerical simulations predicted that the barriers should be functioning perfectly for the measured material properties, with no infiltration into the coarse layer. When pseudo-optimized hydrologic parameters were used in the numerical model, we were able to simulate the barrier failure observed in the experiments, but not the observed differences between the two experiments. Moreover, the measured hydrologic parameters (for Richard's equation) were nearly identical in both experiments, with different observed results. Thus, this exercise did not provide a mechanistic explanation of the actual differences in flow behavior. To explain the observed differences, we measured the surface profiles and roughness of the two sands and conducted an additional vapor diffusion experiment. Based on theoretical calculations and vapor diffusion experiments, the primary controlling mechanism in this system appears to be the magnitude of surface area and roughness of the sediments, resulting in varying rates of infiltration due to differing film flow infiltration velocities.

Hydrologic modeling of unsaturated flow conducted using Richards' equation excludes dry regime flow processes, which can have significant impacts on flow and transport in dry systems in semi-arid and arid climates. Therefore, for experiments conducted at low saturation, considering material characteristics (such as surface area and roughness) in addition to traditional hydrologic properties are necessary to fully describe the system behavior. We believe this is an especially important point to consider when dealing with capillary barrier design. Future numerical models could be improved by extending the hydraulic conductivity- saturation functions to mimic water film infiltration velocities as a function of surface roughness.

Despite the fact that water infiltrated the lower coarse material in both of our experiments, it is notable that the majority of the water was diverted by the drains in both cases (93.3% for the angular sand and 99.7% for the rounded sand). Depending on design requirements, this system may be considered a fully functioning capillary barrier. However, for more stringent design requirements, greater consideration of vapor diffusion and water film infiltration in material selection may be required.

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